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Measurements of NaI:Tl Electron Response using SLYNCI: Comparison of Different Samples

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***Abstract*– This paper measures the sample to sample variation in the light yield non-proportionality of NaI:Tl, and so explores whether this is an invariant characteristic of the material or whether it is dependent on the chemical and physical properties of tested sample.**

In this work we report on the electron response of nine crystals of NaI(Tl), differing in shape, volume, age, manufacturer and quality. The non-proportionality has been measured at the SLYNCI facility in the energy range between 3.5 to 460 keV. The Scintillation Light Yield Non-proportionality Characterization Instrument (SLYNCI) is a next generation Compton Coincidence device, explicitly designed to study the "non-proportionality" of the electron response in scintillators and the contribution of this effect to the intrinsic energy resolution.

We also discuss the gamma response, x-ray excited emission spectra and decay times for the nine crystals, in order to provide a complete characterization of their physical properties and determine whether the mechanism of scintillation varies between samples.

I. Introduction

It has been observed that for most scintillators the luminous efficiency, defined as the number of photon produced per energy unit, depends on the excitation energy. At present, there is general consensus to recognize that this effect, which is known as non-proportionality effect, is a key contributor to the degradation of energy resolution [1]-[4]. In other words, the fundamental limit of the energy resolution for a given scintillator material depends not only on counting statistics but also on its light yield proportionality.

A powerful way to study the electron response of a scintillator is the Compton Coincidence Technique (CCT) introduced by J.D. Valentine and B.D. Rooney, [1]. The basic idea of the CCT is illustrated in fig.1. The crystal under study is coupled to a photomultiplier tube and is irradiated with a well-collimated ^{137}Cs source. When Compton scatter occurs in the scintillator, the scattered electron is then absorbed within the crystal while the secondary gamma is detected in coincidence by means of a high purity Germanium detector. In this way it is possible to relate the amount of light produced in the crystal to the energy of the Compton scattered electron that can be evaluated knowing the energy of the secondary gamma. A wide range of energy, for the electrons generated in the scintillator, can be thus investigated by varying the angle of detection of the secondary gamma rays.

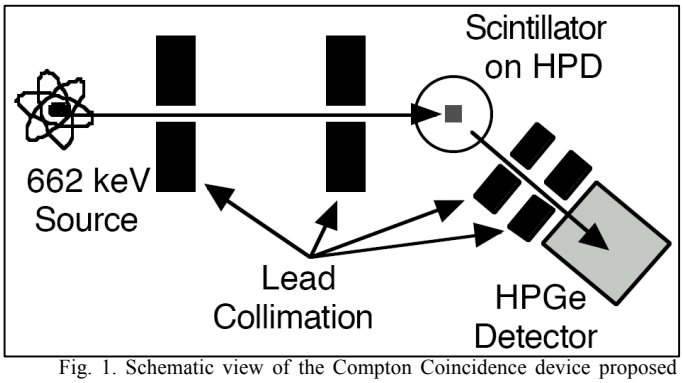

Fig. 1. Schematic view of the Compton Coincidence device proposed by J. Valentine and B. Rooney

The original design of the Compton Coincidence instrument was limited by a very low counting rate, which imposed several weeks of data collection for the characterization of a single sample. For this reason, the CCT method has only been able to measure the electron response of a few scintillator materials [1]-[9], and in particular, only one crystal of each material was studied. Therefore, the question of whether the non-proportionality effect is an inherent, sample-independent property of the scintillator remained to be determined.

The Scintillation Light Yield Non-proportionality Characterization Instrument (SLYNCI) is a second generation Compton Coincidence device that has been installed at the Lawrence Livermore National Laboratory in collaboration with the Lawrence Berkeley National Laboratory [10]. This newly developed facility acquires data roughly two orders of magnitude faster than the original instrument built by Valentine and co-workers, improving dramatically the possibility to apply the CCT for the testing of many scintillator materials and a large number of samples. Detailed systematic studies of electron response can thus now be performed at the SLYNCI facility, in order to gain a deeper knowledge of the nature of the light yield non-proportionality and its effect on the energy resolution.

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II. Equipments and Methods

A. The SLYNCI Facility

A schematic view of the Scintillation Light Yield Non-Proportionality Characterization Instrument is depicted in fig. 2; a detailed description of the instrument is presented in [1]. The SLYNCI facility employs five high purity germanium detectors, a collimated 1mCi Cs-137 source and a highly linear XP2060B Photonis photomultiplier tube, in a much more compact design compared to the original Compton Coincidence instrument. The Cs-137 source irradiates the scintillator under study from a distance of 18 cm, and the HPGe detectors are located 10 cm away from the crystal surrounding it. Each of them subtends an angle of about 30º and thus detects a broad range of Compton scattered gamma energies. The radioactive source is placed on a rotating support so that the angle of the incident 662 keV gamma ray irradiating the crystal can be rotated by 15º. In this way, with two source positions, respectively 0º and 15º relative to the HPGe detectors, it is possible to investigate the electron response of the scintillator in the full energy range between 3.5 to 460 keV.

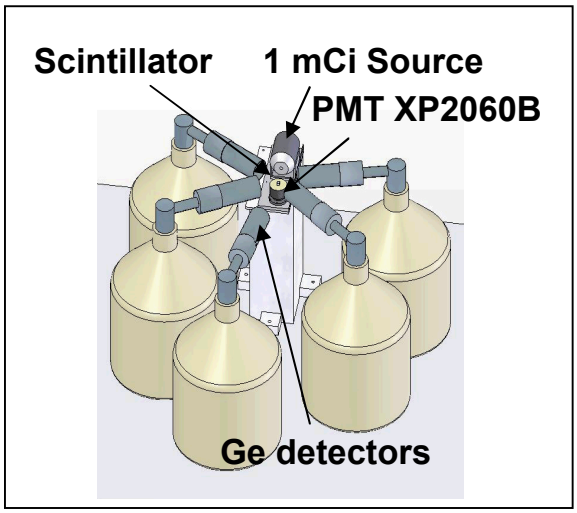

Fig. 2. Schematic view of the SLYNCI

The read-out of the PMT and HPGe detector signals is realized by means of a fully digital data acquisition system, which utilizes two digitizer 6U VME board form Struck Innovative System (SIS). Each board consists of a 6 channel 100 MHz ADC with 12-bit resolution. One advantage of using a digitizer to record the waveforms from the PMT is the possibility to study the scintillation light yield non-proportionality effect for different decay components.

B. The tested NaI(Tl) crystals

In this work we tested nine NaI(Tl) samples, which present differences in shape, volume, age, manufacturer and quality; In table 1 a detailed list of the tested crystals is reported. They include four sets of "paired" crystals (crystals that were procured at the same time from the same vendor, and so should have nearly identical scintillation properties) plus one unpaired crystal.

In particular, we characterized the response of four NaI(Tl) crystals from St. Gobain, of identical shape and volume that were purchased at the same time; for two of them we requested a particularly good (<10% fwhm) energy resolution at 122 keV, while the other two are standard St. Gobain production. We also tested two old Scionix crystals which had yellowed, an old, large Harshaw sample, and finally two new ScintiTech crystals with same shape but different volumes.

TABLE I
THE TESTED CRYSTALS

Sample ID	Shape	Dimensions	Manufacture	Age	Quality
# 568	Cylinder	0.5"Ø× 0.5"	St. Gobain	New	R<10% @ 122keV
# 569	Cylinder	0.5"Ø×0.5"	St. Gobain	New	R<10% @ 122keV
# 570	Cylinder	0.5"Ø×0.5"	St. Gobain	New	Standard
# 571	Cylinder	0.5"Ø×0.5"	St. Gobain	New	Standard
# 572	Cube	1 cm^3	Scionix	Old	Yellowed
# 573	Cube	1 cm^3	Scionix	Old	Yellowed
# 574	Cylinder	1" Ø×1"	Harshaw	Old	Standard
# 578	Cylinder	1 cm Ø×1 cm	ScintiTech	New	Standard
# 579	Cylinder	1 cm Ø×3 cm	ScintiTech	New	Standard

III. The Electron Response

To evaluate the electron response for the nine NaI(Tl) samples under study, we acquired the coincidence events between the PMT and the HPGe detectors at the SLYNCI facility. For each collimator position we acquired 6 million events, including both single and coincidence events, for a collection data time between 10 to 12 hours, depending on the volume of the samples. The acquired single events were used for on-line calibration of the PMT and the HPGe detector positioned directly in front of the collimator; in fact this detector detects the gamma rays scattered at small angles and thus measures the low electron energy, for this reason it is particularly sensitive to gain drift variation.

In fig. 3 the relative light yield for the nine NaI(Tl) crystals under study is reported as a function of the electron energy. The data distributions are normalized at the energy value of 450 keV. In the same plot, the NaI(Tl) electron response measured by J. Valentine and coauthors [1], is shown for comparison.

This plot clearly highlights that there are sample-to-sample variations in the electron response of NaI(Tl) for different crystals. In particular, it is interesting to observe that we measured a 10% variation in the low energy range for sample #568 (round red marker) and #570 (square green marker), which have same shape and volume, and are produced form the same manufacturer. However the light yield data distributions for "paired" crystals, i.e. crystals having the same manufacturer and specifications, consistently match through the whole energy range, as for example for NaI(Tl) #568 (round red symbol) and #569 (triangular light blue symbol). Since the acquisition run for NaI(Tl) #569 was performed two weeks after that of sample #568, the agreement of these data

distributions also highlights the good time stability of the SLYNCI facility.

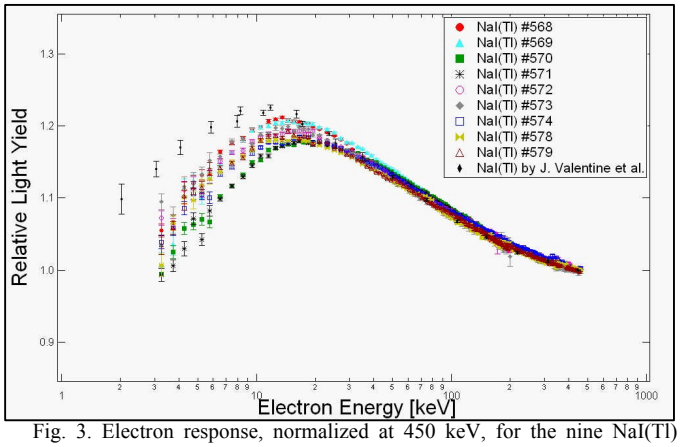

Fig. 3. Electron response, normalized at 450 keV, for the nine NaI(Tl) under study. The NaI(Tl) relative light yield measured by J. Valentine et al. is reported also for comparison

IV. The Gamma-Ray Response

The gamma ray response of the nine NaI(Tl) crystals was tested using five radioactive sources (^{109}Cd, ^{133}Ba, ^{241}Am, ^{57}Co and ^{137}Cs), emitting gamma rays in the energy range between 22.5 keV to 661.657 keV. We acquired these pulse height spectra at the SLYNCI facility with the same experimental set-up employed for the electron response measurements.

In order to evaluate the light yield and the fwhm (full width at half maximum) energy resolution as a function of the gamma ray energy, we performed a Gaussian fit procedure on the main emission peaks of the acquired pulse height spectra to estimate the centroid and the width. For the photopeaks from x-rays emitted by ^{109}Cd and ^{133}Ba, which can not be individually resolved, we applied a weighted average method to determine the centroid and, similarly, the energy of the associated peaks.

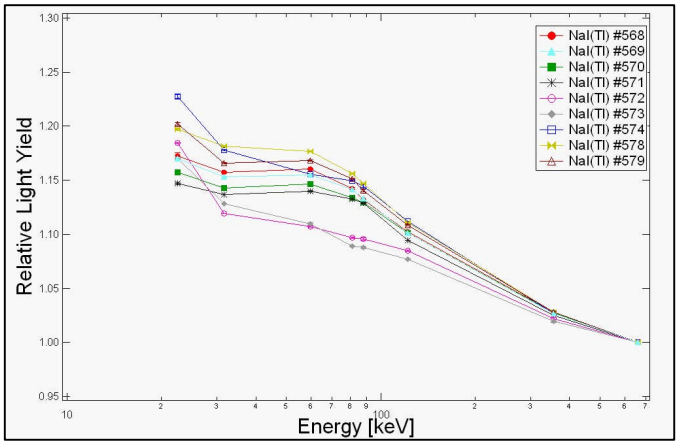

Fig. 4. Relative light yield as a function of the gamma energy for the nine NaI(Tl) samples under study.

Even considering the differences in volume, packaging and coupling to the PMT, for the nine tested NaI(Tl) samples we observed quite large differences in the crystals brightness, up to a factor of ~1.6.

In fig. 4, the measured light yield, normalized at 662 keV, is presented as a function of the gamma radiation energy. As expected we can observe an increase of the light yield with decreasing energy, and the drop of the data distribution around 30 keV corresponding to the Kα edge of iodine.

As we already observed for the electron response, "paired" crystals showed comparable light yield data distributions as a function of the energy.

From the acquired pulse height spectra we also evaluate the FWHM energy resolution of the samples.

At 662 keV, the St. Gobain crystals provided energy resolution of 6.5-6.8%, while for the yellowed Scionix samples we measured a value slightly worse than 11%, clearly demonstrating the degraded quality of these crystals. In fig. 5 the measured energy resolution is reported as a function of the gamma-ray energy.

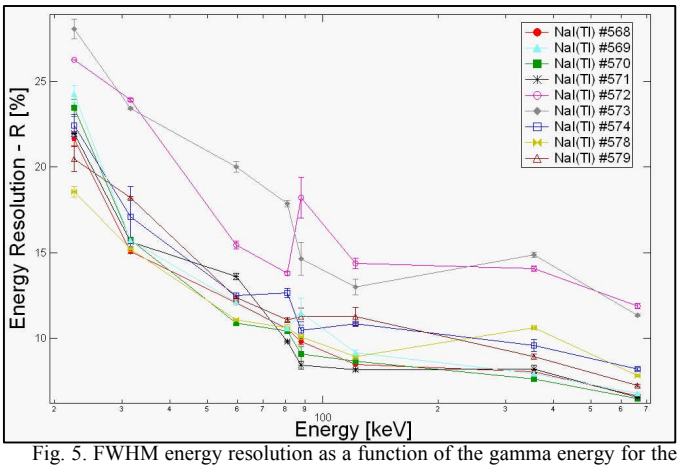

Fig. 5. FWHM energy resolution as a function of the gamma energy for the nine NaI(Tl) samples under study.

V. X-ray Excited Emission Spectra

Emission spectra of the nine NaI(Tl) under study were measured at room temperature, irradiating the samples with an x-ray beam characterized by a mean energy of 30 keV (peak energy at 50 keV) and an intensity of 2kRad/min. The x-ray excited scintillation was collected into a SpectraPro-2150i spectrometer (Acton Research Corp.) coupled to a PIXIS:100B charge-coupled detector (Princeton Instruments Inc.). The CCD is thermoelectrically cooled to 70ºC and has a 1340 x 100 pixel array. For each crystal sample the background is firstly measured with the CCD shutter closed; then we acquired a blue spectrum (between 200 and 360 nm), a green spectrum (between 360 and 620 nm), and finally a spectrum in the red region (between 620 and 1000 nm). More

details on the experimental set-up and procedure are reported in [11].

We observed very similar emission spectra for samples produced by the same manufacturer. In fig. 6 the spectra acquired for three of the tested NaI(Tl) crystals are presented: dotted blue line for sample #568 by St. Gobain, dashed green line for sample #572 by Scionix (yellowed crystal), and full red line for sample #578 by ScintiTech. All the spectra have been corrected for the transmission of the set up. In fig. 5 the pure NaI emission spectrum measured by Moszynski et al. [12], is also presented for comparison.

To evaluate the different components in each spectrum, we performed a multiple Gaussian fit on the data distributions. For the St. Gobain crystals we observed two bands with peaks at 420 nm, corresponding to the thallium emission, and a satellite at 380 nm, presumed to be due to impurities. This component shows an intensity of 20% compared to the main Tl band. The ScintiTech samples show the same two peaks at 420 nm and 380 nm, plus an additional component at 340 nm; the two satellite peaks have intensities equal to 40% and 15% compared to the main band, respectively. For the Scionix yellowed samples the multiple-Gaussian fit estimated the presence of the peaks at 420nm, 380 nm and 340nm and an additional strong emission at 480 nm.

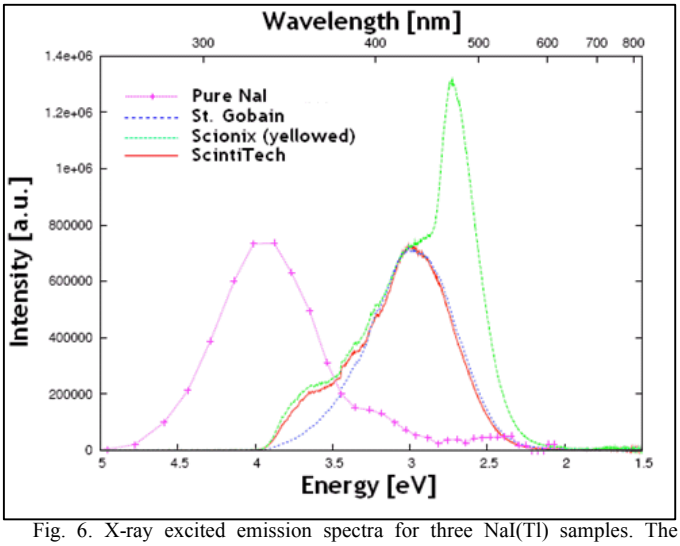

Fig. 6. X-ray excited emission spectra for three NaI(Tl) samples. The emission spectrum for pure NaI as measured by Moszynski et al. (ref. [12]) is reported for comparison.

VI. Decay Time Measurements

The decay time measurements were performed by irradiating the NaI(Tl) crystals with an x-ray beam of mean energy of 19 keV. The x-ray gun runs at 30kV and a 0.51 mm thick Aluminum foil is used to reduce the presence of soft x-rays in the beam. Details on the experimental set-up are reported in [13].

Fig. 7 shows the decay signal acquired for NaI(Tl) sample #568, as an example. In order to estimate the decay time from the acquired signals we performed a fit procedure on the data distribution with the function:

$$I(t) = \frac{A_1}{T} \cdot \exp(-t/T) + \frac{A_2}{T - T_r} \cdot \left[\exp(-t/T) - \exp(-t/T_r)\right]$$

This fitting function takes into account two different mechanisms for the excitation of the Thallium ions: the first component represents the prompt excitation, while the second component represents the delayed one.

For all of the tested NaI(Tl) crystals we measured nearly identical values for the prompt decay time T (typically 173 ns) and the rise time T_r (59 ns). However, we observed large differences in the ratio between A_1 and A_2. As an example, for NaI(Tl) #568 we measured a ratio of 59.5% for the prompt component, while it becomes more significant for NaI(Tl) #578, being the 79% of the total decay signal.

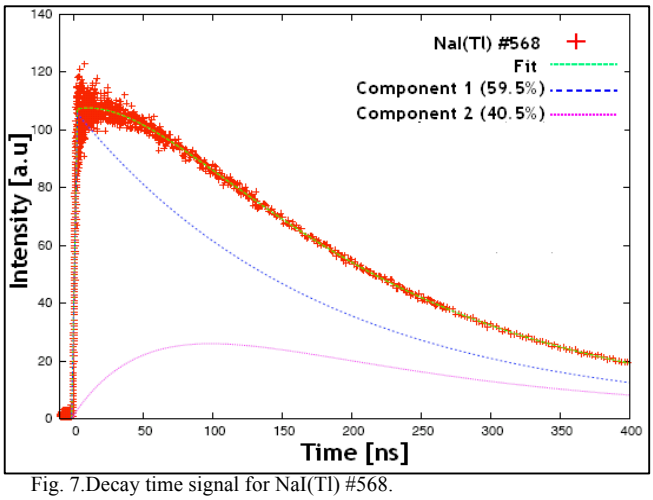

Fig. 7.Decay time signal for NaI(Tl) #568.

VII. Conclusions

The electron response, measured over nine NaI(Tl) samples with different characteristics, showed a sample-to-sample variation up to 10%. We furthermore observed large differences in the gamma ray light yields, x-ray excited emission spectra and ratio of the decay components on the tested crystals. However "paired" crystal pairs from the same manufacturer, which could originate from the same boule or at least have been grown under the same conditions, evidenced nearly identical properties.

Our collaboration is working to develop a fundamental predictive theory of the electron non-proportionality in order to explain and to understand the origins of the observed differences in the measured electron responses. To this aim we are particularly interested to combine the electron response, gamma response measurements, and decay time measurements, and relate them to the differences in chemical and physical properties of the scintillator materials using the SLYNCI facility.

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